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Project Summary

A Mathematical Model for a Fluidized-Bed Coal Gasifier

M.J. Purdy, R.M. Felder, and J.K. Ferrell

A devolatilized Kentucky bituminous coal and a New Mexico subbituminous coal have been gasified with steam and oxygen in a pilot-scale fluidized-bed reactor. The reactor was operated at pressures of 570-840 kPa (80-120 psia), molar steam-to-carbon feed ratios of 0.6 to 1.9, and average bed temperatures of 795-1010°C (1460-1850°F). The coal feed rate ranged from 14 to 33 kg/hr (30-73 lb/hr).

The experimental results provided a basis for the formulation and evaluation of mathematical models of the gasifier. A simple three-stage gasifier model and a two-phase bubbling-bed gasifier model were developed. The simple model assumes instantaneous devolatilization of coal at the top of the fluidized bed, instantaneous combustion of carbon at the bottom of the bed, and steam/carbon gasification and water gas shift reaction in a single perfectly mixed isothermal stage. The bubbling-bed model compartmentalizes the reactor into an incipiently fluidized emulsion phase and a solids-free bubble phase. The solids are assumed to be well-mixed as in the simple model, but plug flow of the gases is assumed. Model options allow for consideration of a jetting region at the gas inlet of the bed and elutriation of fines.

Parameters were estimated with each model for each of the two feedstocks. The main difference in the optimal values was in char reactivity, for which that of the New Mexico coal was roughly an order of magnitude greater than that for the Kentucky char. Using the optimal parameter values, the models were used for all experimental runs.

Model predictions and experimental results showed good agreement in all cases. Taking into account departures from isothermality in the bed, jetting at the gas inlet, and particle size distributions in the feed coal did not provide significant improvements in the ability of the model to correlate the data, except for elutriation rates, which are highly sensitive to the concentration of fines in the feed. Sulfur gas formation is better predicted by equating sulfur and carbon conversion than by using published kinetic correlations.

The effects of various operating parameters and phenomena on reactor performance were determined using the models. As expected, carbon conversion and make-gas production both increase with bed temperature, steam-to-carbon feed ratio, and solid-phase space time. Both also go up with pressure; but, above about 1.7 MPa, the increases are negligible. At the temperatures studied, the water/gas shift reaction falls short of equilibrium for pressures lower than 2.1 MPa (confirming experimental results), but the reaction is close to equilibrium at pressures above this value.

This Project Summary was developed by EPA's Industrial Environmental Research Laboratory, Research Triangle Park, NC, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).

Introduction

As a part of a continuing research program on the environmental aspects of fuel conversion, the U.S. EPA has sponsored a research project on coal gasification at North Carolina State University (NCSU). The overall objective of the project is to characterize the

gaseous and condensed-phase emissions from the gasification/gas-cleaning process, and to determine how emission rates of various pollutants depend on adjustable process parameters.

The facility used for this research is a small coal-gasification/gas-cleaning pilot plant, including a continuous fluidized-bed reactor, a cyclone separator, a venturi scrubber, and absorption and strippng towers and a flash tank for acid gas removal. Process control, data acquistion, and data logging systems, and an extensive analytical laboratory complete the facility.

In experiments conducted to date, a devolatilized Western Kentucky bituminous coal, a New Mexico subbituminous coal, a North Carolina peat, and a Texas lignite have been gasified with steam and oxygen. The experimental results are summarized in several EPA reports.

The primary function of the NCSU gasification reactor is to provide a reproducible synthesis gas for studies of the potential environmental impact of coal gasification processes. The development of correlative and predictive models of the gasifier was felt to be an indispensable adjunct to planning and implementing the overall experimental program. As part of the study, two gasifier models were developed and used to correlate the data for gasification of the bituminous char and the New Mexico subbituminous coal—one simple, the other relatively complex. This report summarizes the principal features of both models, and reports the model correlations of the gasification data for both feedstocks studied.

Model Description

The three-stage well-mixed reactor model and the bubbling-bed model are described.

Three-Stage Well-Mixed Reactor Model

The first stage of the modeling studies was to formulate the simplest possible model which incorporates the principal gasification reactions and the gross physical characteristics of the reactor, and to determine the degree to which the gasifier performance could be correlated by this model. A well-mixed fluidized-bed model was used for this purpose. The model assumes instantaneous devolatilization and oxidation, followed by gasification in a perfectly mixed bed. The model takes as input the average reactor bed temperature and pressure; the bed dimensions; feed rates of coal, steam, oxygen, and nitrogen; solids holdup in the bed; and ultimate analysis of the feed coal. It calculates carbon conversion and make-gas flow rate and composition.

Devolatilization is presumed to occur instantaneously where the coal enters the bed. The extent of devolatilization and composition of the volatile products are taken from rapid pyrolysis data obtained in a separate study at NCSU and additional data for a western subbituminous coal close in composition to the one used in this study.

After devolatilization has occurred, the following reactions are presumed to take place:

$$C + H_2O = CO + H_2 \quad (1)$$
$$C + 2H_2 = CH_4 \quad (2)$$
$$2C + H_2 + H_2O = CO + CH_4 \quad (3)$$
$$CO + H_2O = CO_2 + H_2 \quad (4)$$
$$C + 1/2O_2 = CO \quad (5)$$
$$C + O_2 = CO_2 \quad (6)$$
$$S + H_2 = H_2S \quad (7)$$
$$H_2S + CO_2 = COS + H_2O \quad (8)$$

Reactions 5 and 6, the oxidation steps required to supply heat for the remaining reactions, are assumed to occur instantaneously in a zone of negligible volume separate from the gasification zone. All oxygen in the feed gas is taken to be consumed to form CO and CO_2, with the combustion product distribution being governed by the relation:

$$C + aO_2 = (2\text{-}2a)CO + (2a\text{-}1)CO_2$$

where a, the combustion product distribution coefficient, is an adjustable model parameter. A value of a = 0.5 indicates that all CO is formed, while a = 1.0 indicates that only CO_2 is formed.

Reactions 1, 2, and 3 are the reactions with which the Institute of Gas Technology correlated gasification kinetics data. These kinetic expressions, summarized in the body of the report, include as an adjustable parameter a char reactivity coefficient, f_o, which has values ranging from 0.3 (for low-volatile bituminous coal char) to about 10 (for North Dakota lignite).

Reaction 4 is the water/gas shift reaction. A model option allows the assumption of shift reaction equilibrium or the use of kinetic rate equations. The rate expression used contains an adjustable parameter, f_{wg}, which accounts for the varying catalytic activities of different chars.

The rate of Reaction 7 is estimated by assuming that the sulfur conversion equals the carbon conversion, and that all converted sulfur forms hydrogen sulfide by Reaction 7. The flow rate of COS is then determined by assuming equilibrium of Reaction 8.

In summary, the model has three adjustable parameters: the intrinsic char reactivity factor, f_o; the CO/CO_2 combustion product distribution parameter, a; and the water/gas shift reactivity factor, f_{wg}. The other coefficients of the various rate and equilibrium equations are prescribed by the model. Details of these equations and of the model computational procedure are given in the report.

Bubbling-Bed Model

The two-phase bubbling-bed model divides the reactor into two main sections: a bed region, and a freeboard region above the bed. Consideration of a jetting region at the gas inlet of the bed is a model option that can add a third section to the model.

The NCSU gasifier uses a top coal feeding system, with the feed particles falling through the hot exiting gas stream before entering the bed. In the freeboard region, the particles (heated by the gas stream) are dried and devolatilized. In addition, the fines in the coal feed stream may be eluted by the exiting gas stream. The model of the freeboard section accounts for all of these phenomena.

If elutriation is considered, the amount of fines blown out of the reactor is first calculated by assuming that all particles with terminal velocities less than the gas velocity at the top of the reactor are eluted. The blowover solids are assumed to be devolatilized at the gas exit temperature.

After the blowover calculation, the flow rate and size distribution of the remaining coal are determined. The devolatilization products for this stream are calculated, assuming equilibrium yields at the temperature at the top of the bed. The devolatilization products are assumed to be evenly evolved along the length of the freeboard region.

Heat transfer from the gas to the coal particles is calculated for each class of particle sizes using a single-sphere heat transfer correlation. It is assumed that there are no radial temperature gradients within particles. The latent heat of devolatilization is neglected. The heat of vaporization of moisture is included in the model, with an average value of 37.7 kJ/gmol being used for all conditions. It is assumed that the heat required to vaporize the moisture in the feed coal is taken from the gas phase.

Particles are assumed to have constant volumes in the freeboard region, so that the particle densities decrease as drying and devolatilization proceed. No gas-phase reactions are considered in this

region. The model contains an option to consider a jetting region at the gas inlet of the bed. The jet penetration height and jet angle are calculated using earlier correlations.

The gases in the fluidized bed are assumed to be in plug flow, while the bed solids are assumed to be well-mixed with respect to composition and size. Mass transfer between the jet and emulsion phases is determined using a mass transfer coefficient correlation.

The region between the jet penetration height and the top of the bed is assumed to be a two-phase fluidized region, consisting of an incipiently fluidized emulsion, and a solids-free bubble phase. Both gas phases are assumed to be in plug flow, while the solids are assumed to be perfectly mixed throughout the jet and fluidized-bed regions. The assumption of well-mixed solids throughout the bed is supported by experimental results. Mass transfer rates between the two phases are calculated using earlier correlations.

For both the jet and fluidized regions, the temperature may be determined three ways: (1) a specified isothermal value; (2) an imposed temperature profile; and (3) a profile calculated assuming adiabatic reactor operation. For all, the solids and gases at any height are assumed to be at the same temperature.

The following reactions are assumed to occur in the emulsion phase:

$$C + H_2O = CO + H_2 \quad (9)$$
$$C + 2H_2 = CH_4 \quad (10)$$
$$2C + H_2 + H_2O = CH_4 + CO \quad (11)$$
$$C + O_2 = CO_2 \quad (12)$$
$$2C + O_2 = 2CO \quad (13)$$
$$S + H_2 = H_2S \quad (14)$$
$$CO + H_2O = CO_2 + H_2 \quad (15)$$
$$2CO + O_2 = 2CO_2 \quad (16)$$
$$2H_2 + O_2 = 2H_2O \quad (17)$$
$$CH4 + 2O_2 = CO_2 + 2H_2O \quad (18)$$
$$H_2S + CO_2 = COS + H_2O \quad (19)$$

The jet and bubble phases are solids-free, so that only Reactions 15-19 may occur.

Reactions 9-11 are gasification reactions, with rates given by the previously discussed kinetic expressions. Carbon combustion Reactions 12 and 13 are described by an earlier rate equation. The combustion rate expression yields the rate of carbon consumption, but does not specify the gaseous products. The combustion product split between CO and CO_2 is therefore calculated as outlined in the context of the simple three-stage model. The CO/CO_2 distribution coefficient, a, can be determined from an earlier correlation or left as an adjustable parameter.

Gas phase combustion occurs according to Reactions 16-18. The reactions are very fast, and are assumed to go to instantaneous completion. If oxygen is insufficient for complete combustion, the reactions probably compete for oxygen at different rates. However, the reactions are expected to have a small overall effect on the model predictions. So, for simplicity, the three reactions are assumed to compete equally for available oxygen.

The water/gas shift reaction is considered to occur in both the bubble (jet phase in the jetting region) and emulsion phases. In the emulsion phase, the reaction occurs predominantly on the char surface, which acts as a catalyst. A model option also allows for the assumption of shift reaction equilibrium.

Hydrogen sulfide formation by Reaction 14 can be calculated two ways: by using earlier kinetic rate equations, or by assuming equality of the sulfur and carbon conversions with all sulfur being converted to H_2S. The latter assumption is based on experimental data.

The elutriation rate from the bed is determined following the size reduction calculations. The size distributions of the elutriated solids and the bottoms exit solids are then calculated, and the solids elutriated from the bed are added to the solids blown out at the top of the reactor to form the total elutriate flow.

Details of the computational procedure for the model are given in the report. Also given are tabulations of the physical property data and correlations used in formulating both models.

Results

Results are given separately for the well-mixed stage model and the bubbling-bed model.

Well-Mixed Stage Model

The coal reactivity coefficient, f_o, the combustion product distribution coefficient, a, and the water/gas shift reactivity parameter, f_{wg}, were evaluated by using a Pattern Search routine to minimize a function of the sum of squared deviations between predicted and measured values of gasifier performance variables, including carbon conversion, dry make-gas flow rate, and mole fractions of hydrogen, carbon monoxide, and carbon dioxide in the make-gas. The runs with the best mass balance closures were chosen to provide the data base for the parameter estimation. For the Kentucky bituminous char, the parameter values obtained were:

$$f_o = 0.276$$
$$a = 0.828$$
$$f_{wg} = 1.29 \times 10^{-5}$$

The value of a, given above, indicates that 66% of the carbon oxidized forms CO_2 and 34% forms CO. An earlier equation predicts a = 0.57 at 760°C and a = 0.52 at 1100°C, while several gasification studies have assumed a = 1.0.

The value of $f_{wg} = 1.29 \times 10^{-5}$ indicates that the shift reaction rate is approximately five orders of magnitude less than the rate typically obtained in catalytic shift reactors. An earlier study used a shift reactivity value of 1.7×10^{-4} in modeling the gasification of a bituminous coal by the Synthane process. The larger earlier value many be attributed to differences in the coals used in the studies.

Using the optimal parameter values, the model was run for all char runs. Plots of predicted vs measured values of carbon conversion, dry make-gas flow rate, and sweet gas heating value are shown in Figures 1-3. (The sweet gas is defined as the dry make-gas with the CO_2 and H_2S removed.) The close proximity of the points to the 45 degree line is gratifying in view of the simplicity of the model.

The same optimization procedure used to determine the optimal parameters for char was used to find the optimal parameter values for the New Mexico subbituminous coal, using fast pyrolysis product data to calculate the devolatilization gas product distributions. The resulting parameter values were:

$$f_o = 4.20$$
$$a = 1.00$$
$$f_{wg} = 1.2 \times 10^{-4}$$

The char reactivity parameter for coal is much larger than that obtained for the Kentucky char. The combustion coefficient does not differ greatly for the coal and the char; however, the shift reactivity parameter is an order of magnitude greater for the coal than for the char. A possible reason for the greater shift reactivity of the New Mexico coal (compared to the char) is the different ash contents of the two materials: the coal had a feed ash content of 22.6%, while the ash content of the char was only 10.7%. The minerals in the ash fraction of coal serve as catalysts for the shift reaction. In addition, during gasification, the coal was converted to a greater extent than the char, giving an even larger difference in ash contents. The shift reactivity parameter obtained in this study for a subbituminous coal agrees well with the earlier value used for a bituminous coal ($f_{wg} = 1.7 \times 10^{-4}$).

The model was used to correlate the New Mexico coal run data using optimal parameter values. In general, the agreements between model predictions and

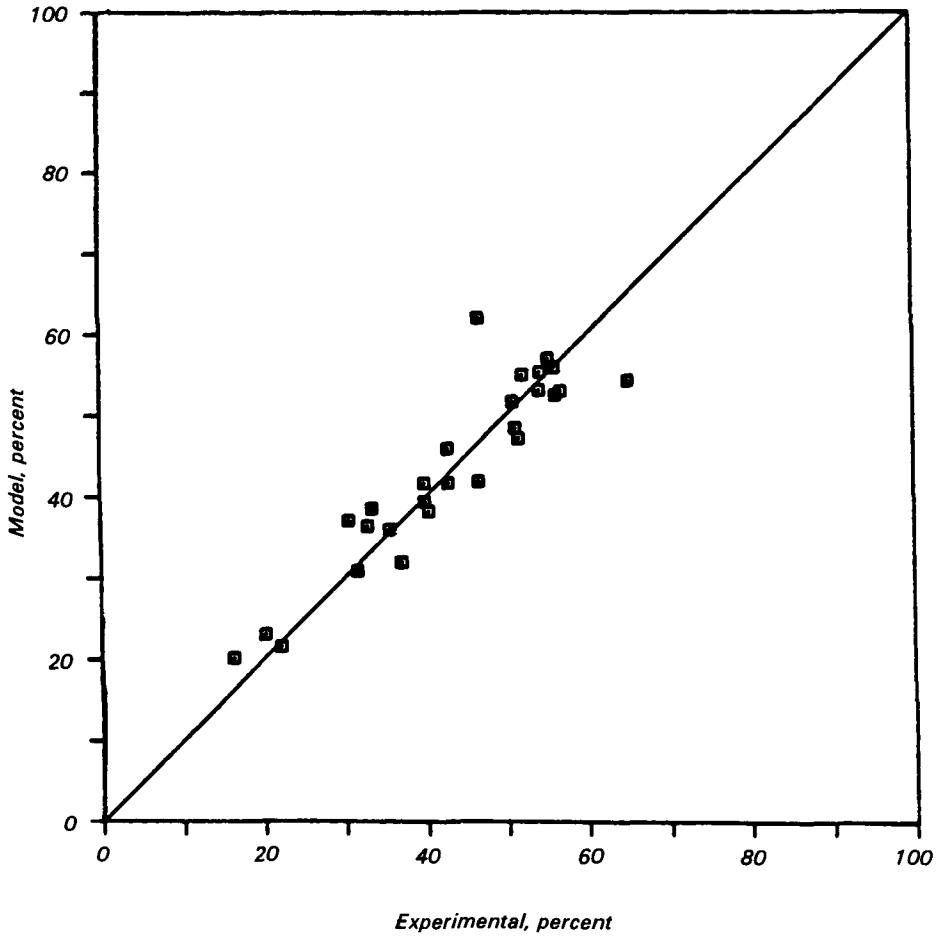

Figure 1. Predicted vs. experimental carbon conversion for Kentucky char, well-mixed model.

experimental measurements were excellent. Comparisons of predicted and measured reactor variables are shown in Figures 4 and 5.

For each run, the water/gas shift equilibrium ratio was calculated from both experimental measurements and model predictions of the gas composition. A plot of the predicted vs experimental values of this ratio is given in Figure 6. The substantial degree of scatter may be attributed to the simplicity of the model, and equally to the fact that the mole fractions which are the constituents of K_{wg} are interdependent, so that an experimental error in one of them affects the values of the others.

The significance of this plot emerges when it is compared with Figure 7, which shows the values of K_{wg} predicted assuming shift equilibrium. This assumption leads to overprediction of K_{wg} and lends support to the conclusion that the shift reaction should not be assumed to proceed to equilibrium.

Bubbling-Bed Model

The two-phase bubbling-bed model of the gasifier contains the same three parameters as the well-mixed model. In addition, a fourth parameter—the char abrasion constant (K_a)—is required to calculate the particle size reduction due to abrasion in the fluidized bed. All are properties of the material being gasified. The parameters are independent of processing conditions, and (once determined) are applicable to any set of reactor conditions.

The two-phase bubbling-bed model of the gasifier is much more complex than the well-mixed model. It also requires a much larger program and more computer time for completion. As a consequence of the model's size and limited available computing time, the optimization procedure was less detailed than that for the well-mixed model. To speed the optimization computations, the step sizes used in searching for the optimal set of parameter values were increased from those used in optimizing the well-mixed model parameters, and smaller data bases were used.

The optimal parameter values for the bituminous char were

$$f_o = 0.80$$
$$a = 1.00$$
$$f_{wg} = 1.5 \times 10^{-4}$$
$$k_a = 3.0 \times 10^{-4}$$

The values of f_o, a, and f_{wg} are all higher than the corresponding values determined for the well-mixed model. The differences may be due to the gas/solid contacting assumed in the two models. In the two-phase model, a large fraction of the gas passing through the bed is in the bubble phase and does not contact the coal. The well-mixed model does not allow for such gas bypassing, and as a consequence may underpredict the reactivity parameter values.

Using the optimal parameter values, the model was run for the conditions of all Kentucky char runs for which acceptable mass balance closures were obtained. The model predictions for carbon conversion and gas production are plotted versus the experimental results in the report. The plots show good agreement between the model predictions and the experimental measurements, but similar to and not significantly better than those shown in this summary for the well-mixed stage model.

Parameter estimation for the New Mexico subbituminous coal yielded the following results:

$$f_o = 8.60$$
$$a = 1.00$$
$$f_{wg} = 2.2 \times 10^{-4}$$
$$k_a = 4.5 \times 10^{-4}$$

The char reactivity factor for the New Mexico coal is roughly 11 times greater than that for the Kentucky char. This agrees with the experimentally observed trend, and with the results of the well-mixed model (f_o ratio of 15). The combustion product coefficient is the same as that determined for the Kentucky char, and indicates that CO_2 is the sole product of the carbon/oxygen reaction. Similar values were obtained for the shift reactivity factor, while the greater abrasion constant value for the New Mexico coal indicates that it is more friable than Kentucky char.

Using the above values, the model was used to correlate the results for New Mexico coal. Parity plots of the main reactor performance variables are shown in the report. As for the bituminous char correlations, good agreement was obtained between the model predictions

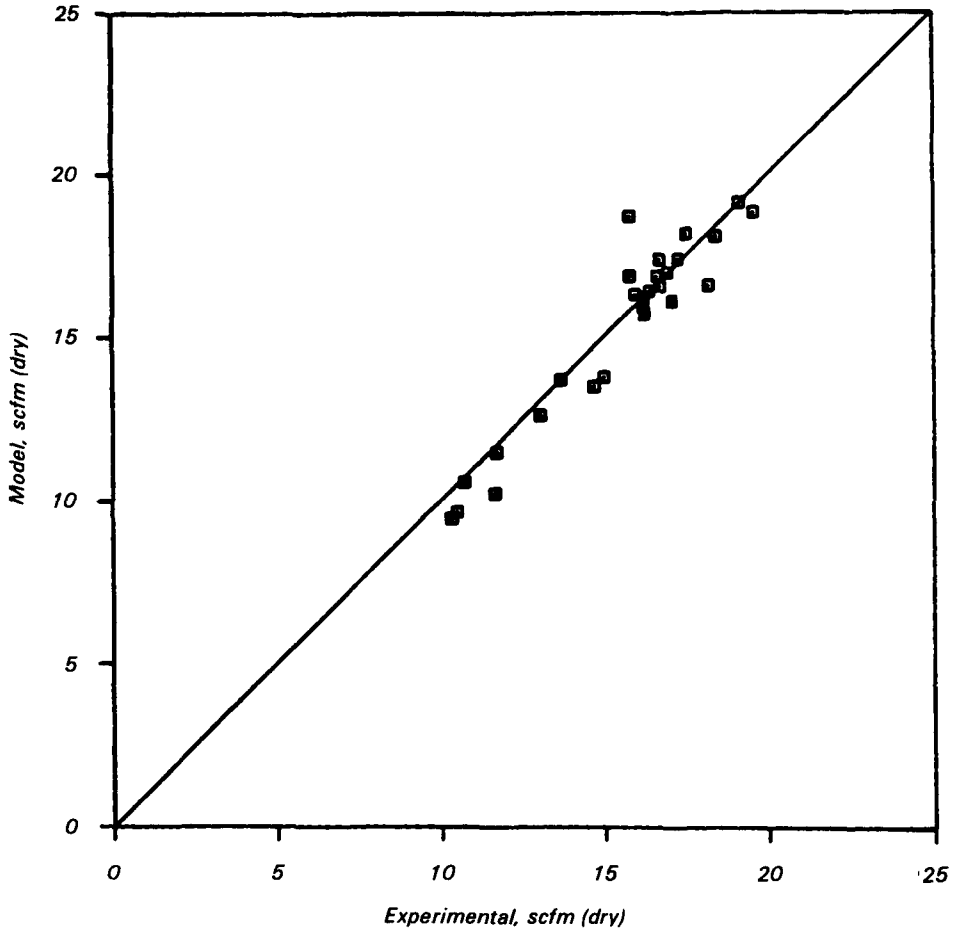

Figure 2. Predicted vs. experimental gas production rate for Kentucky char, well-mixed model.

and experimental results, but not significantly better than with the simpler model.

Quantitative measures of goodness of fit for both models lead to the conclusion that no significant improvement in data correlation is obtained by resorting to the more sophisticated bubbling-bed model —certainly not enough to justify the considerably greater computation time required to implement the latter model.

Studies were performed to determine the effect on reactor performance of an axial temperature profile in the bed; jetting at the gas feed nozzles; and particulate elutriation. For all, it was found that relatively little error in calculated carbon conversion and dry make-gas flow rate is introduced if these phenomena are neglected. Sulfur gas formation is better predicted by equating sulfur conversion and carbon conversion than by using published kinetic correlations.

The well-mixed stage and bubbling-bed models were run for several sets of hypothetical reactor conditions to determine the effects of selected operating variables on reactor performance. The variables examined were temperature, pressure, H_2O/C feed ratio, solid-phase space time, and feed particle size distribution.

The results are presented in detail in the report. As expected, carbon conversion and gas production increase with bed temperature, steam-to-carbon feed ratio, and solid-phase space time. Both also increase with pressure; but, above a pressure of about 1.7 MPa (250 psia), the increases are negligible. Another noteworthy result is that the water/gas shift reaction falls short of equilibrium for pressures lower than 2.1 MPa (300 psia), as earlier results indicate, but the reaction is extremely close to equilibrium at pressures above this value.

The bubbling-bed model takes the feed particle size distribution into account, whereas the well-mixed model does not require size distribution. The two-phase model was run for several feed size distributions to determine their effect on reactor performance predictions. All operating conditions were held constant except feed size distribution. Results show that feed size distribution has a dramatic effect on the elutriation rate, but a relatively small effect on carbon conversion and gas production. The make-gas shows a decrease in the CO/CO_2 ratio with increasing particle size. The hydrogen content in the dry gas increases with increasing particle size.

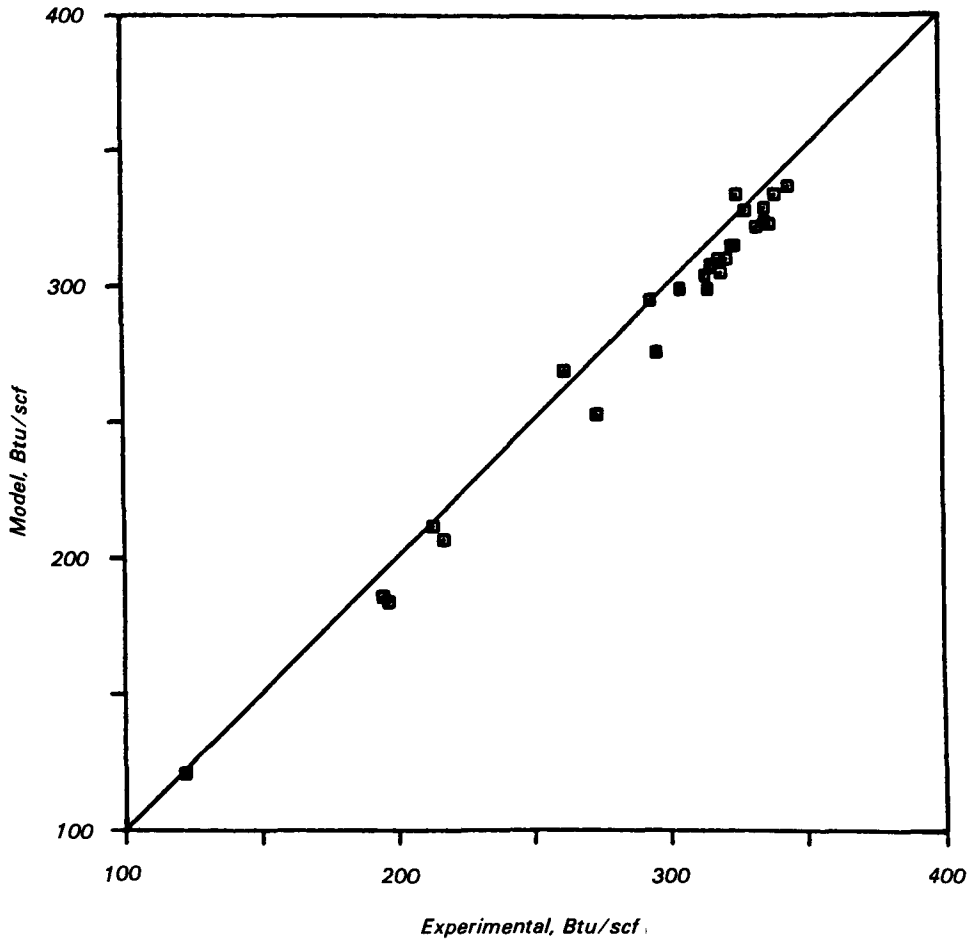

Figure 3. Predicted vs. experimental heating value for Kentucky char, well-mixed model.

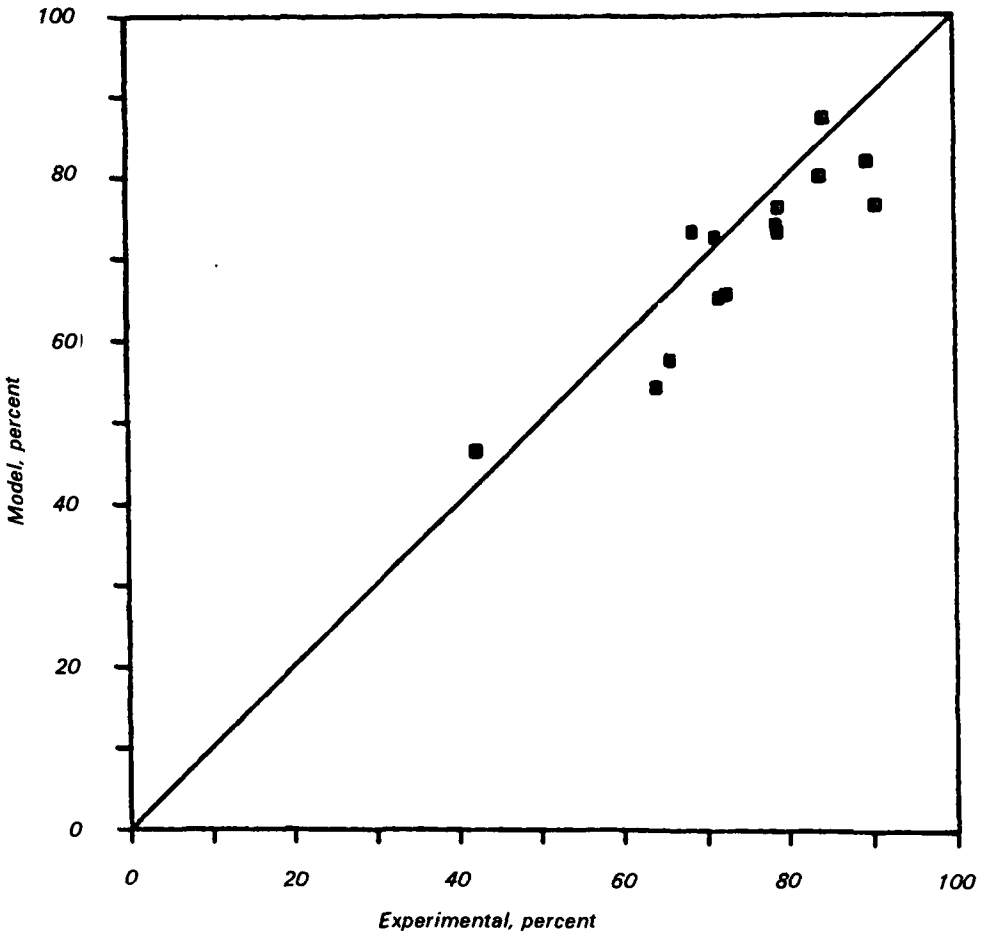

Figure 4. *Predicted vs. experimental carbon conversion for New Mexico coal, well-mixed model.*

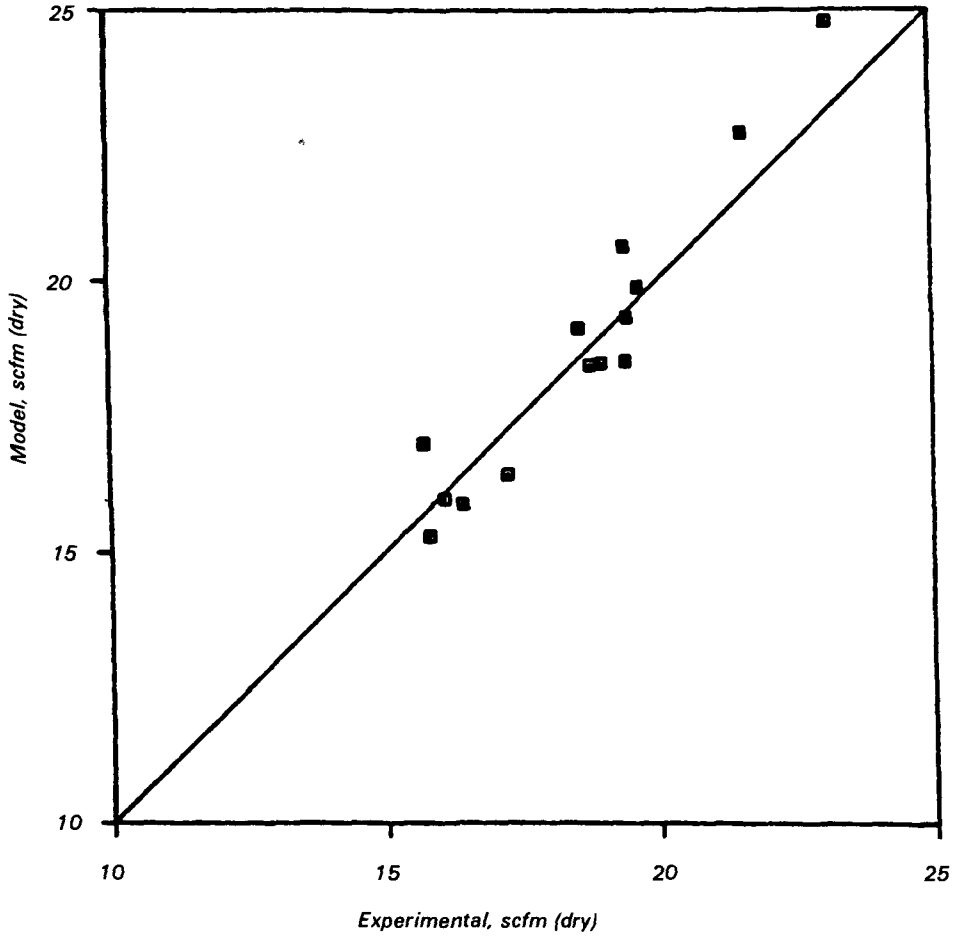

Figure 5. Predicted vs. experimental gas production rate for New Mexico coal, well-mixed model.

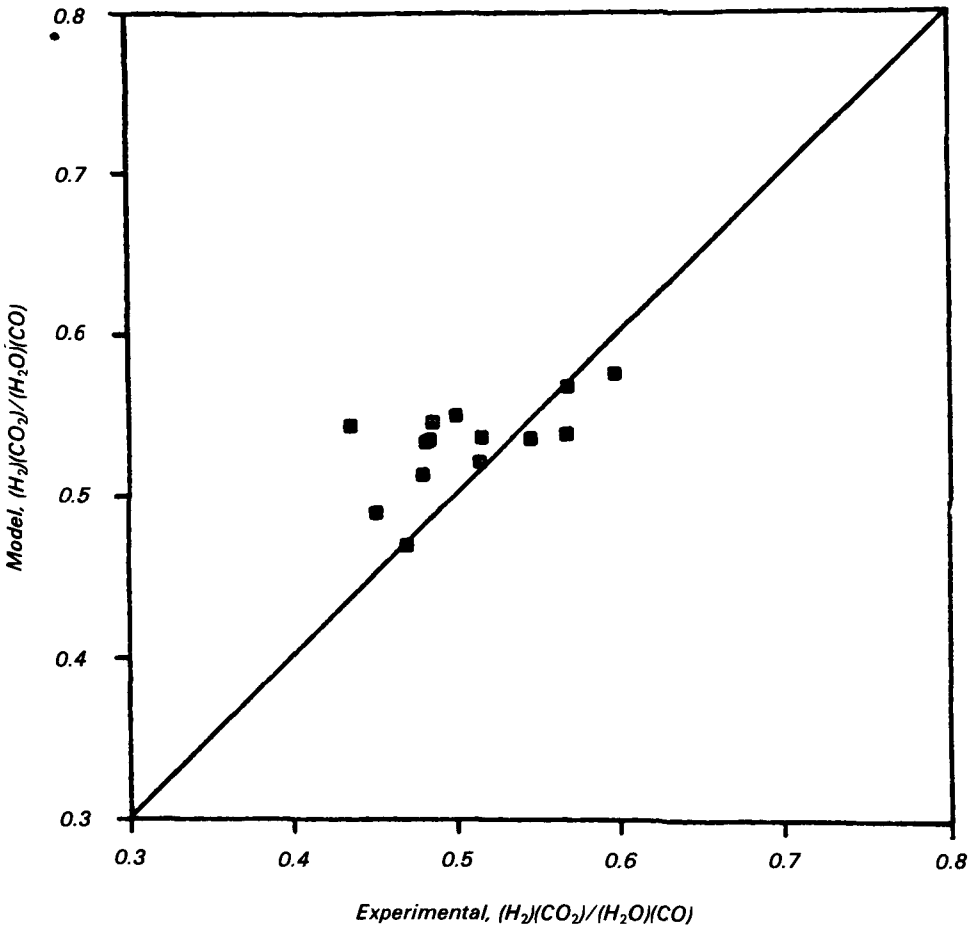

Figure 6. *Predicted vs. experimental K_{wg} value for New Mexico coal, well-mixed model.*

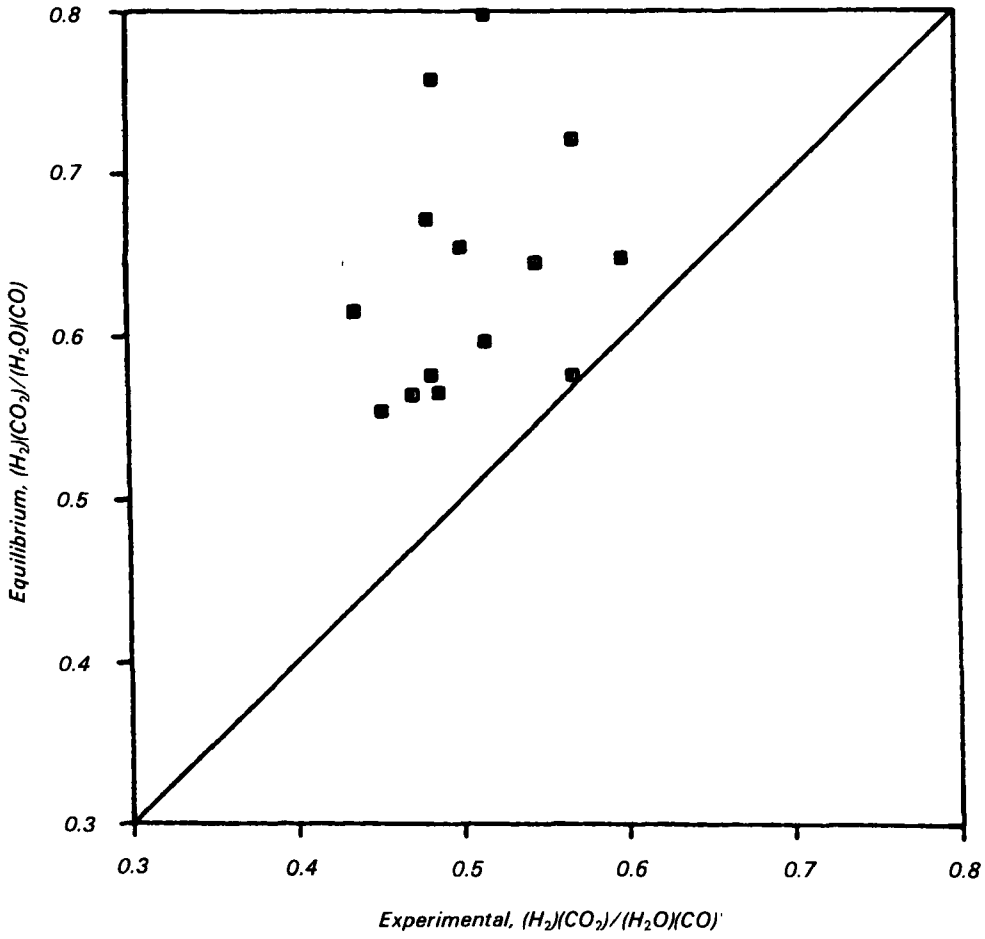

Figure 7. *Equilibrium vs. experimental K_{wg} value for New Mexico coal.*

M. Purdy, R. Felder, and J. Ferrell are with North Carolina State University, Raleigh, NC 27650.
***N. Dean Smith** is the EPA Project Officer (see below).*
The complete report, entitled "A Mathematical Model for a Fluidized-Bed Coal Gasifier," (Order No. PB 84-209 469; Cost: $20.50, subject to change) will be available only from:

National Technical Information Service
5285 Port Royal Road
Springfield, VA 22161
Telephone: 703-487-4650

The EPA Project Officer can be contacted at:

Industrial Environmental Research Laboratory
U.S. Environmental Protection Agency
Research Triangle Park, NC 27711

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Information
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